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Submitted to:

<http://lib-www.lanl.gov/la-pubs/00796402.pdf>

CHARACTERIZATION OF RADIOACTIVE AND HAZARDOUS WASTE AT LOS ALAMOS NATIONAL LABORATORY

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ABSTRACT

Radioactive and hazardous waste from actinide processing in nuclear facilities must be characterized in order to ensure safe and regulatory compliant disposal. Nondestructive assay techniques are used to determine nuclear material content and analytical chemistry methods are used to establish composition, but these activities are time-consuming and expensive. Regulations allow acceptable knowledge to be used in order to reduce analytical requirements, provided the integrity of documentation can be demonstrated. The viability of the program is based upon record management and traceability and must withstand the rigors of audit. Electronic inventory and data-gathering systems are implemented to reduce record management and reporting burdens.

Keywords: actinide, waste, characterization, data, assay

1.0 INTRODUCTION

Los Alamos is one of the United States of America's (USA's) National Laboratories owned by the United States Department of Energy (DOE) and operated by the University of California (UC). The laboratory is one of the original Nuclear Weapons Complex Laboratories dating back to Project Y of the Manhattan Engineering District during World War II. Consequently, research with radioactive materials has been conducted at Los Alamos for over half a century and remains one of the primary responsibilities of this institution. The Nuclear Materials Technology (NMT) Division is responsible for the operation of two nonreactor nuclear facilities: the Chemistry and Metallurgy Research (CMR) Building in TA-3, and the Plutonium Facility at TA-55.

The activities at these two nuclear facilities generate a variety of waste types from sanitary and salvage to radioactive, hazardous [as defined by the Resource Conservation and Recovery Act (RCRA)] and mixed (radioactive and hazardous) wastes. All of these materials must be properly segregated and handled safely, and in accordance with a variety of rules and regulations governing the treatment, packaging, identification, storage, transportation and disposal of each specific type of waste.

Compliance with a multitude of regulations must be demonstrated to internal and external audit teams evaluating the integrity of NMT's waste management program. Record keeping must be robust, and correlation of records with the waste must be demonstrated from initial identification of an item as waste to final disposition. A number of analytical and expert knowledge techniques from disparate sources are used to characterize the composition of these waste streams. Reports must be generated on a regular basis for external use by disposal facilities and oversight organizations and for internal use by management and environmental professionals. By their very nature, regulations are constantly changing and invariably require more controls to be implemented and additional information to be collected in order to achieve full characterization of the waste. This tends to drive up costs which, considering the static or declining budgets of recent times, results in less money for programmatic and mission activities. This situation, in turn, leads to delays in schedules and frustrated sponsors.

A major focus of waste management has been to reduce analysis requirements, simplify data gathering, and automate report generation in the face of rapidly changing regulations and ever-increasing demands for data collection. Automated, computer-based inventory systems and hand-held bar-code scanners have provided the inspiration for developing an integrated system to tie together container identification and characterization information with records and report generation. These efforts are beginning to come to fruition by offsetting additional reporting requirements through saving personnel time on the operations floor and through clerical and data-entry personnel. Improvement in data integrity and quality assurance gains are also being obtained by automating many calculations and data transfer operations that have historically resulted in data transcription errors as a result of manual data entry and manipulation.

This paper presents the evolution and recent innovations in the waste characterization and waste management process in NMT Division at the TA-55 Plutonium Facility and CMR Building.

2.0 LOS ALAMOS NATIONAL LABORATORY

Los Alamos National Laboratory is situated on the Pajarito Plateau on the eastern flanks of the Jemez Mountains at an elevation of 2,200 m. It is about 40 km northwest of Santa Fe, the capital of the state of New Mexico, in the USA's desert southwest. The Laboratory occupies about 111 km² in Los Alamos County, and the climate is classified as high desert, receiving about 30 cm of rainfall annually. The Laboratory employs about 10,300 University of California employees and contractors. Operations are overseen by the DOE's Los Alamos Area Office. The Laboratory is organized geographically into 40 Technical Areas (TAs) east and south of the town site. The Nuclear Materials Technology Division is one of 18 divisions at the Laboratory organized in specialized scientific areas of expertise. NMT Division occupies all buildings at TA-55, known as the Plutonium Facility and, one structure at TA-3, the Chemistry and Metallurgy Research Building.

3.0 NUCLEAR MATERIALS TECHNOLOGY DIVISION

The NMT Division is comprised of 760 personnel organized into 12 Groups at TA-55 and the CMR Building. These groups are organized into six infrastructure and facilities groups and six operations groups. The resources and operations in these groups are formed into project teams in support of programs. The mission of NMT Division is primarily one of support for stockpile stewardship activities for the USA's nuclear arsenal. These activities include a wide spectrum of actinide research and development tasks, component surveillance, special nuclear material (SNM) recovery, pit fabrication, and the supporting operations that these activities require, such as analytical chemistry, nondestructive assay and waste management. Nondefense programs are also supported, for instance, the fabrication of plutonium-238 heat sources for radioisotopic thermoelectric generators for the National Aeronautics and Space Administration.

The Plutonium Facility is a heavily reinforced, two-story concrete structure with a top operations floor divided into four wings and a service floor located underneath. Many safety controls are engineered into the facility that was designed in the aftermath of the 1969 Rocky Flats fire. Negative-pressure, stainless-steel glove boxes are the primary barrier protecting workers from nuclear materials. Engineered barriers are supplemented by standard radiological control practices, access control and monitoring activities. The Plutonium Facility was commissioned in 1978. The older (1952) CMR building is much larger in size compared with the plutonium facility and was constructed in eight wings (two are administrative) around a spinal corridor. The wings are divided into three floors with services in both the basement and attic and operations on the ground floor. Radiological controlled areas are in the centers of the wings with office and nonradioactive laboratory space on the outside walls. Filter towers are located at the ends of the wings, opposite the spinal corridor. With the exception of the hot cell operations, most activities in CMR Building including analysis and metalography handle much smaller quantities of nuclear materials than those activities conducted in the Plutonium Facility. Most waste is therefore of lower activity than that from the Plutonium Facility.

4.0 WASTE CLASSIFICATION

Operations in NMT Division produce a wide variety of waste types. Radioactive wastes from actinide processing operations are contaminated with radioisotopes from defense materials and their daughter products. By definition, transuranic (TRU) waste: (1) is contaminated with radioisotopes with atomic numbers greater than 92, (2) decays by alpha decay, (3) has half-lives greater than 20 years and 4) has an activity greater than 3.7 kBq/g. Typically, TRU waste comes from glove-box operations and some maintenance activities on highly contaminated process systems. TRU and mixed-TRU waste is destined for disposal by deep geologic emplacement at the Waste Isolation Pilot Plant (WIPP) in Carlsbad, New Mexico.

Wastes contaminated with these isotopes (below the 3.7 kBq/g threshold) are disposed of as low-level waste. These wastes are generated inside radiological control areas, but outside of the glove-box lines. Low-level waste is packaged for on-site disposal by burial in shallow trenches. The low-level Waste Acceptance Criteria (WAC) is less stringent than for WIPP, but ensures that the disposal facility remain in conformance with the limits established by the performance assessment (PA) mandated by siting regulations.

LANL does not deal with high-level waste because there are no spent fuel reprocessing activities at the Laboratory. High-level waste problems in the defense arena are principally the concern of the DOE sites that operated production reactors and separated special nuclear materials from targets (Hanford Site in Richland, Washington and Savannah River Site outside of Aiken, South Carolina).

Hazardous waste is defined in RCRA, which was passed into law in 1976. These waste materials are defined by regulation: to exhibit a hazardous characteristic (flammability, corrosiveness, reactivity, or toxicity) or are listed by name on one of four lists (F, K, U, and P). The issue of DOE compliance with these environmental regulations was subject to much litigation in the early 1990s before an agreement was reached between the DOE and the Environmental Protection Agency (EPA) in the form of the Federal Facilities Compliance Agreement (FFCA). Operations at the Laboratory are currently under regulation by the New Mexico Environment Department, and considerable staff time is spent in maintaining the Laboratory's Part B Operating Permit under RCRA. Hazardous wastes must be treated to very exacting standards under the regulation in order to dispose of them by burial (Land Disposal Restrictions).

Sanitary solids are taken to the county landfill; sanitary liquid goes to an on-site treatment plant. Recycle efforts have been implemented to reduce the quantity of waste in the sanitary solid waste stream by recycling materials such as white paper and cardboard. Metal recycle and salvage operations that result in the sale of discarded equipment and furniture also play key roles in waste minimization.

5.0 WASTE REGULATIONS

A number of regulations impact waste management in NMT Division. The Atomic Energy Act of 1954 (AEA) and its amendments form the basis for control of SNM. NMT Division has an elaborate system of Special Nuclear Material Control and Accountability (NMC&A) built around the Materials Accounting and Safeguards System (MASS). Operations are organized into a number of Material Balance Accounts (MBAs) in order to track the use of nuclear materials. Accounts are balanced on a quarterly basis and SNM loss through the waste stream is an important consideration.

DOE Order 435.1, "Radioactive Waste Management," defines DOE's expectations on how radioactive wastes are to be managed within the DOE Complex. Descriptions are given for provisions designed to protect the public, workers, and the environment from radioactive waste operations. General guidance is provided on waste: definition, management, characterization, certification, waste acceptance, documentation, facility design, storage, and monitoring. The concise order is supplemented with voluminous guidance documents.

RCRA is applicable to hazardous or mixed wastes as defined previously and supplements radioactive waste disposal regulations. RCRA regulations are very prescriptive, requiring precise marking and labeling and configuration of storage areas. Specialized waste storage areas are designated as: less-than-90-day storage, satellite storage, universal waste storage and treatment, storage and disposal (TSD) areas. These storage areas must be maintained and inspected in the defined manner. Facilities that produce hazardous or mixed waste must have a permit and TSDs must be defined in the permit. Specific operations that produce hazardous waste must be defined in the permit. Much emphasis is placed upon comprehensive characterization of waste items with identification of the hazardous components in the waste.

The Toxic Substances Control Act (TSCA) regulates wastes containing polychlorinated biphenyls (PCBs) and asbestos. Remediation must be performed in a prescribed manner by certified personnel in protective clothing. Wastes must be packaged and labeled properly, and disposal must be in dedicated disposal facilities.

The Hazardous Materials Transportation Act (HMPT) applies to all waste packages because they are transported over public thoroughfares to get to the TSD sites. Packaging must be compatible with the waste and designed to provide the prescribed level of containment. Again, the contents of the waste must be accurately known in order to properly categorize the material for marking, labeling, attaching placards, and producing shipping papers. Packages are defined from "strong-tight" through "type B" designed for maximum transportable quantities of nuclear materials. Extensive Safety Analysis Reports for Packaging (SARPs) are made for unique shipping package designs and are licensed by the Nuclear Regulatory Commission (NRC).

These are a sampling of the major regulations that help define the waste management program and drive characterization requirements for wastes generated in NMT Division. Further exploration of the regulations in this context would be counterproductive.

6.0 WASTE CHARACTERIZATION

Complete characterization of waste items generated from NMT operations is critical to waste program compliance. In order to be cost effective and efficient, waste must be segregated at the source to the extent possible by waste type. This works to minimize the most problematic waste types; those that are expensive to

treat and dispose or those for which no disposal path exists (indefinite storage commitment). An example of this is the mixture rule under RCRA that states that a little hazardous waste mixed with a lot of nonhazardous waste results in lots of hazardous waste (dilution is not the solution to pollution!).

Comprehensive waste characterization, as required by the aforementioned regulations, is expensive and time consuming. Expensive sampling and analysis depletes limited funds and regulations set time limits on disposal to discourage waste abandonment. Merely following the regulations is a substantial commitment for regulatory professionals who have a working knowledge of the regulations, regular inspections to verify regulatory compliance, maintenance of equipment and storage areas, support of continual program modification to keep abreast of regulatory changes, and maintenance of an integral quality assurance program. Waste acceptance criteria, environmental regulations, and transportation regulations set very rigid expectations for waste constituents to be disclosed. In the case of RCRA, expectations for sampling and analysis are very prescriptive and define requirements such as chain of custody, holding times for volatile organic compounds, sampling techniques, and quality control (EPA SW-846, "Test Methods for Evaluating Solid Waste"). Sampling for hazardous chemical constituents alone can run into thousands of dollars per sample, and complicating the process by introducing radioactive materials (mixed waste) can easily escalate analytical costs.

Fortunately, EPA allows for the use of process knowledge, termed acceptable knowledge (AK), to narrow the sampling and analysis requirements. AK must be thorough and defensible, though. AK guidance is provided in the EPA document: "Waste Analysis at Facilities that Generate, Treat, Store and Dispose of Hazardous Waste." NMT has spent considerable time collecting and organizing the information needed to build AK files on various waste streams. Information is gathered by interviewing process operators (past and present), and from checking operating logs, process engineering flow diagrams, operating procedures, purchase orders, feed stream analysis, and actual sample results. In this manner, sampling can be limited to the regulated constituents of interest.

NMC&A required by the AEA of 1954 has a major impact upon waste operations, as discussed previously. It is also very important that disposal sites know the radioisotope content of the waste sent to them for emplacement. Repositories must be constructed in sites in accordance with regulations that define expected isolation of the waste from the environment over the design life of the facility (10,000 years as defined by 10 CFR 960, "General Guidelines for the Recommendation of Sites for Nuclear Waste Repositories." It is expected that the designers of the repository can demonstrate, through computer simulation (PA), the successful performance of the repository. One of the limits of the model is radioisotope inventory. These limitations are defined in the WAC for the generators that are sending waste for disposal. Generators must be able to demonstrate the accurate radioisotope content of the waste. DOT regulations also require that radioisotope content be known to within 95% of the total isotope inventory by activity.

7.0 ACCEPTABLE KNOWLEDGE

An emphasis is placed upon the use of acceptable knowledge in order to narrow the scope of analysis for RCRA hazardous constituents. The undertaking of documenting AK was considered from a historical perspective because of the large quantity of retrievably stored waste onsite that was emplaced during the 1970s and 1980s. Approximately 9,000 m³ of TRU waste in 200-L drums and boxes were stored in this manner and are currently being recovered by the Transuranic Waste Inspectable Storage Project (TWISP). This is a sizeable quantity of waste that will need to be certified and characterized for compliance with the WIPP WAC for disposal in Carlsbad, New Mexico. This project will benefit from NMT AK because most of the waste had its origin in NMT operations.

The AK effort has the advantage of detailed historical records that document which waste items were produced by the specific processes for NMC&A purposes. These records provide information on waste generating process status (PS) that narrow the point of origin to a specific chemical process and a geographic location. Operations in the Plutonium Facility yielded about 175 PSs. Some are long-term operations, and others were of a temporary nature as research and development activities that no longer exist. A number of PSs that produced waste of the same characteristics were grouped together to form seven quantifiable waste streams for AK purposes: (1) nitrate operations, (2) chloride operations, (3) pyrochemical operations, (4) special operations, (5) metal operations, (6) miscellaneous operations and (7) heat-source operations.

In addition to NMC&A requirements pertaining to waste disposal, two pivotal events resulted in greater complexity for the management of waste and records keeping. The first was the establishment of a repository for TRU waste disposal in Carlsbad, New Mexico, and implementation of the WIPP WAC in 1980. The second was the FFCA that required federal facilities to comply with federal environmental regulations and to identify,

segregate, treat, store and dispose of hazardous waste in accordance with RCRA (1992). Both of these events resulted in substantial increases in characterization and record keeping for waste.

Waste stream acceptable knowledge summary reports are pointer documents that tie together sources of detailed information describing the waste stream characteristics [1]. Descriptive and historical information is presented to provide a basis of understanding because many of these processes are interrelated and very dynamic. The reports begin by identifying common characteristics of the waste stream and identifying those processes that made that type of waste. A description of how the waste is then categorized, segregated, and packaged is discussed, placing emphasis on the constituents of interest, i.e., radioisotopes and any RCRA-regulated hazardous materials present. It is critical to the exercise that the contents of specific waste packages be identified and traced back to the processes that made the waste in order to demonstrate knowledge of the components.

Very detailed discussions are provided for radionuclide content that address products from radioactive decay and the variation in concentration in the residues of various separations and recovery processes. RCRA-regulated constituents are identified, starting with characteristic waste (reactivity, flammability, corrosivity and toxicity), and proceeding through commonly occurring toxic heavy metals to volatile and semivolatile organic materials (VOC & SVOC). In some cases, headspace gas analysis of actual waste drums was used to verify process knowledge. Other items of interest, such as WAC prohibited items, were discussed, including absence of explosives, free liquids, compressed gases, large sealed containers, etc.

A variety of methods were used to collect information for the summary reports; existing documents were examined for information regarding currently recognized waste streams, sources of information on waste content were identified and cataloged, personnel involved in the waste management process were interviewed and the processes that generated the waste were thoroughly investigated. These sources of information are identified in the summary reports in tabular form, titled as the Acceptable Knowledge Roadmap. Process flow diagrams of key processes were also simplified and included as attachments to aid in understanding processes and material flows. Samples of waste documentation records are also attached to provide examples of waste records available and their comprehensiveness.

8.0 NONDESTRUCTIVE ASSAY OF WASTE

Waste is assayed at several points in the disposal process. Initially generators use the knowledge of their activities to make the decision if an item is a recoverable residue or is a waste item. This determination is based upon the quantity of SNM in the item, its weight and matrix. The SNM content is expressed in terms of grams of SNM per kilogram of waste matrix. Studies were conducted to evaluate the practical aspects of material recovery and safeguards & nonproliferation issues, as well as economics given the current surplus situation. Guidelines are made available to generators on which to base the waste determination under this Plutonium Disposition Methodology (PDM) [2]. Guidance is organized by material matrix (plastics, graphite, metal, glass, etc.) and SNM Material Type (MT) which defines the isotopic composition of the nuclear material. The waste item is bagged out of the glovebox line after visual inspection by waste management technicians and sent for nondestructive assay.

Segmented gamma scan is used to assay waste items of light (low Z) matrices that transmit decay gamma rays with minimal self-attenuation. Collection of 129.3 kilo electron volt (keV) and 417 keV gamma rays are used to determine the quantity of plutonium-239 present in the sample. MT is used to infer the quantity of other plutonium isotopes present. Operations in the plutonium facility use materials of well-known isotopic composition (MT) and rarely mix materials of different material types. Knowledge of the consistency of the isotopic ratios and the nature of the operations conducted allows for accurate determination of isotopic content without resorting to full-spectral analysis. This information is captured on the computerized Waste Management System (WMS) for each waste item.

Waste matrices that are heavy (high Z) or contain uranium-235, must be assayed using neutron coincidence or passive/active neutron interrogation techniques because of high attenuation of the decay gamma rays of interest. Passive systems capture spontaneous fission neutrons to determine nuclear material content in the waste, where as active systems induce fission in the nuclear material and use die-away techniques to determine SNM content. NMT uses the technique that yields the most accurate results, based upon the MT and waste matrix present in the waste (see figure 1).



Figure 1: Passive/active neutron interrogation of TRU waste drum.

Sensitivity and accuracy are very important in determining the radioactive waste category of items. NDA must be able to discriminate between low-level and TRU waste. The WIPP WAC requires that SNM values be reported with twice the uncertainty (2σ). Sensitivity is problematic, considering the relatively high background radiation levels in the Nondestructive Assay Laboratory that is located inside the Plutonium Facility, providing an opportunity for future research and development. Currently, confirmation NDA measurements are conducted at the waste staging and disposal site under low radiation background conditions.

9.0 WASTE DOCUMENTATION AND TRACKING

The quality of waste documentation and correlation with disposal packages are fundamental to program integrity. Information must be accurate and readily retrievable for audit purposes. Waste produced in accordance with the LANL waste management program is accepted for WIPP disposal based upon program certification by the disposal facility established by periodic program assessment.

The management of large amounts of waste information has historically taken a significant effort and presented quality assurance challenges to the program. Information is gathered from multiple sources (generators, waste management technicians, nondestructive assay personnel) and must be compiled and converted for use. These activities introduce opportunities for errors in transcription and calculation. The value of automated data collection and manipulation was recognized in the 1980s, and projects were undertaken to create the computerized system necessary to automate waste management data collection and manipulation. The first system was the computerized Waste Management System (WMS) for TRU waste information. This program has been in use for six years and has eased the administrative burden on both waste management technicians and staff.

The most recent effort, the Waste Information and Tracking System (WITS), was initiated to gather characterization and tracking information on waste other than TRU [3]. It is based upon the use of hand-held palm pilots (Symbol® SPT 1700) that are used in the field for data entry on active screens and tracking of waste packages by use of built-in bar-code readers (see figure 2). Information in the palm pilots is uploaded through custom cradles that link them with personal computers (for data synchronization). The database (Oracle 8®) resides on an Oracle Web server that can be accessed remotely by personal computers (PCs) or notebook PCs. These data can be readily accessed by staff from their PCs for report generation, trending, cost accounting, waste tracking, waste minimization, and research activities.



Figure 2: Field inventory of low-level waste boxes with palm-pilot.

This system is being tested on the low-level, compactable room trash stream. It is especially valuable in providing information needed to complete shipping manifests according to Department of Transportation regulations. Individual 0.056 m³ boxes are combined into DOT Type-A over-packs for shipment to the on-site compaction and disposal facility. Proper marking and labeling of the shipping container as well as the shipping manifest is dependent upon the content of the individual waste boxes. Use of WITS saves data transcription and calculations necessary to prepare these waste shipments. The transfer of data to the disposal facility database can also be accomplished by electronic data transfer, saving additional transcription time and manual quality assurance checks.

10.0 CONCLUSION

NMT Division constantly strives to reduce waste disposal costs while enhancing the integrity of waste operations and maintaining compliance with growing regulatory-driven characterization requirements. Automated data collection and processing using palm pilots and a WEB server database, facilitate this process and meets legal requirements for generator affirmation and disclosure. The challenge in transitioning to electronic format is to retain the familiar data forms and to tailor hardware and software to existing operations. In this manner, waste management personnel are better able to adapt to the new protocol, and impact to on-going operations is minimized. The system must also be designed to readily accommodate the inevitable changes in waste operations while maintaining system integrity and meeting software quality assurance expectations.

Use of process knowledge to eliminate unnecessary sampling and analysis is cost effective, but involves considerable initial effort to document properly. AK takes advantage of existing records and leverages upon systems that provide information on the waste stream even though they may not have been originally intended to do so. These summary documents and files are subject to audit and must be accurately written in accordance with overall program quality assurance requirements.

Successful waste management programs are dynamic; anticipating the future program direction required to meet tomorrow's regulatory needs with sufficient advance notice to accomplish the changes in a manner that does not adversely impact operations.

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